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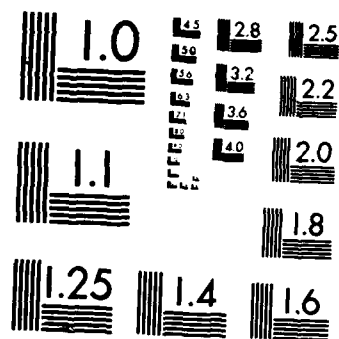
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Signal Processing with Degenerate Four-Wave Mixing (AFOSR-84-0277)

Principal Investigator: George Stegeman, Optical Sciences Center, University of Arizona

We have made progress in a variety of areas within this program since our original report of DFWM in a thin film waveguide.[1] That original experiment utilized liquid carbon disulphide (CS_2) as the nonlinear mixing medium in the form of a cladding on a planar sputtered 7059 glass waveguide. A number of obvious deficiencies exist in configuration:

(a) the physical form of the nonlinear medium (ie a liquid is not very practical),

(b) the optical field strength in the nonlinear medium is small since only the evanescent tails of the guided modes exist in the nonlinear cladding, and

(c) the magnitude of the nonlinear index in CS_2 $n_2 \sim 10^{-18}$ m^2/w is not particularly high.

To improve upon these deficiencies what is required is a nonlinear material preferably in a practical solid form which can be formed into low loss optical waveguides. Semiconductor doped glasses were identified as an excellent candidate material to meet these requirements. This is a glass host containing mixtures of CdS and CdSe compound semiconductor crystallites. We performed bulk DFWM and photoluminescence measurements[2] on these materials and determined that they have a large nonlinear index of refraction ($n_2 \sim 10^{-14}$ m^2/w with fast response times ($\tau < 16$ psec) required for high speed optical signal processing.

Of the number of processing techniques which exist for producing optical waveguides from glass based materials, ion-exchange is one of the most flexible and easiest to implement. We have now established a very successful potassium (K^+)/sodium (Na^+) ion-exchange technique[3] for producing low-loss (few dB/cm) planar waveguides. This process is also amenable to producing channel waveguide structures and devices which we are also undertaking.

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We are currently assessing these semiconductor-doped glass waveguides for application to this waveguide DFWM program. One area of concern is the effect of the ion-exchange process on the nonlinear response of the material since we are not achieving the expected response from waveguide samples which, previous to processing, were showing large DFWM signals in bulk form. Whilst working on this problem we are also developing waveguides utilizing the semiconductor-doped glass as the nonlinear substrate material. These are produced by RF sputtering of a higher index glass film onto the nonlinear substrate to produce the waveguide. Very low-loss waveguides have been produced by sputtering of PSK-2 glass onto the pre-polished color-filters and experiments are in progress to attempt waveguide DFWM in this similar geometry to the CS_2 case but with a higher nonlinearity medium surrounding the guiding layer.

The other major avenue of research pertaining to this program is the use of nonlinear organic materials for waveguide optical signal processing applications. A number of reports of large and fast responding nonlinearities have been made recently and we are currently testing materials of this form from the Celanese Corporation. We spin thin films of company propriotory organic materials onto fused silica and glass substrates and have achieved relatively low-loss guiding (~ 1dB/cm). The coupling process is achieved by using physical relief gratings on the substrate since prism coupling can seriously damage such mechanically soft films.

Other sources of nonlinear organic materials are also being pursued (SUNY Buffalo; JPL) and the possibility of producing nonlinear organic films by deposition in our Langmuir-Blodgett tank is still in our plans. We have succeeded in producing excellent films of cadmium arachidate from single to several hundred monolayers thick. Sources of nonlinear organic LB molecules are now being sought.

1. "Degenerate four-wave mixing in planar CS_2 covered waveguides," C. Karaguleff, G. I. Stegeman, R. Fortenberry, R. Zanoni, and C. T. Seaton. Applied Phys. Letts., **46**, 621 (1985).

2. "Ultrasfast carrier and grating lifetime in semiconductor-doped glasses," S. S. Yao, C. Karaguleff, A. Gabel, R. Fortenberry, C. T. Seaton, and G. I. Stegeman, Appl. Phys. Letts., **46**, 801 (1985).

3. "Waveguide fabrication in nonlinear semiconductor doped glass," C. N. Ironside, J. F. Duffy, R. H. Hutchins, W. C. Banyai, C. T. Seaton, and G. I. Stegeman, Proceedings IOOC/ECOC '85, p. 237.

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Personnell supported on program:

G. I. Stegeman: principal investigator

C. T. Seaton: co-investigator

J. V. Moloney: co-investigator

N. Finlayson: post-doc

A. Gabel: student

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Degenerate four-wave mixing in planar CS₂ covered waveguides

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Degenerate four-wave mixing in a thin-film waveguide is demonstrated for the first time. A reflectivity of 10^{-9} at a power density of 5 MW/cm^2 was obtained for liquid CS$_2$ placed on top of a glass waveguide.

Degenerate four-wave mixing (DFWM) is a phenomenon of considerable interest because of its applications to optical signal processing.[1] Nonlinear signal delay lines,[1] time reversal of optical pulses,[2] optical computing,[3] bistability,[4] and bandpass filtering[5] are among the processing operations that have been discussed. The DFWM signal efficiency is proportional to the product of the electric field amplitudes (power densities) of the three input optical beams overlapping within the nonlinear medium. Guided waves provide an ideal vehicle for DFWM since a guided wave optimizes the power density for a given total beam power. To date, this idea has been demonstrated via several different approaches: in multimode optical fibers,[6,7] within CS$_2$ filled stainless steel capillary tubes,[8] as well as in surface plasmon configurations.[9] Specifically in the area of integrated optics, convolution and time reversal have both been suggested[10] and cross sections have been derived.[11] In this letter we report what we believe is the first demonstration of degenerate four-wave mixing in a thin-film waveguide.

Figure 1 shows the sample and beam interaction geometry used in our experiments. The waveguide consists of a $\simeq 1$-μm film of Corning 7059 glass deposited on a BK-7 substrate. Three high index coupling prisms are arranged on the free-film surface so that the three input guided wave pump and probe beams would intersect. A small quantity of CS$_2$, which has a large (relative to glass) third order nonlinearity $\chi^{(3)} \simeq 2 \times 10^{-20}$ m^2/V^2, was used as a nonlinear cover medi-

um over the part of the waveguide where the beams intersect. The liquid was held in place by a hollowed out glass cell whose bottom surface was well polished and optically contacted to the thin-film surface. After careful alignment of the three coupled input beams, the glass cell is inserted, gently pressed onto the waveguide via a set screw, and filled from the top with CS$_2$. Thus, the evanescent tails of the guided beams in the CS$_2$ generate the nonlinear source polarization. The efficient coupling of three independent beams with prisms was found to be very difficult to achieve and resulted
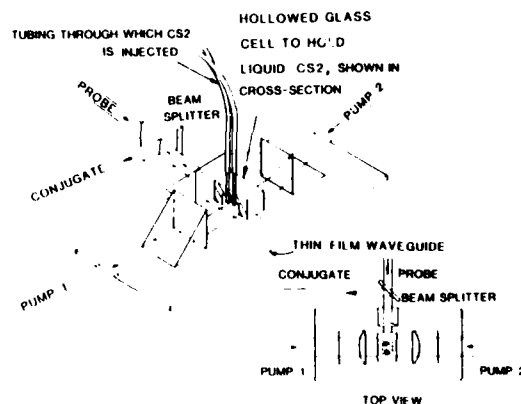

FIG. 1. Sample and coupling geometry showing the thin-film waveguide (Corning 7059 sputtered onto a glass microscope slide), the arrangement of high index coupling prisms, and the glass cell for holding CS$_2$. The probe beam intersects the pump beams at 90° under the liquid cell.

in reduced coupling efficiencies, of the order of 1%.

The salient feature of this guided wave approach is the enhanced field amplitudes of the guided beams at the cover-film interface which can be maintained over centimeter distances due to the tight confinement of the beam within the thin film. Additionally, greater spatial bandwidth (variable angle θ) is available than in fiber optic configurations. Unfortunately, due to the inherent waveguide scattering losses of a few dB/cm, propagation distances and thus interaction lengths must be kept much smaller (on the order of a centimeter) than for fibers.

A passively Q-switched, frequency doubled ($\lambda = 0.53$ μm) Nd$^+$:yttrium aluminum garnet laser was used in the experiments. Energies of 20 mJ per pulse, 10 pulses per second, with 15-n pulse widths, were available from the laser. Due to poor coupling efficiencies (under 1%), peak power densities achieved within the waveguide were only on the order of 100 MW per cm^2 in each beam, and of the order of 6 MW/cm^2 in the CS$_2$ which carried only 6% of the guided wave power. Despite giving a smaller interaction region than a more collinear geometry would provide, a 90° probe angle (relative to the pump beams) was selected in order to minimize background contamination of the signal due to scattering light from the pump beams.

Data collected from the *pin* photodiode monitoring the laser output (power in), and from a photomultiplier tube which measures the conjugate (power out), are stored in a microcomputer for analysis. Measurements were taken for a series of input powers both with all the beams present, and with one of the beams blocked off. This of course defeats the DFWM process, and provides assurance that any observed nonlinearity is not due to detector saturation.

The ideal method to confirm DFWM would have been to directly compare phase and amplitude profiles of the incident probe and emitted conjugate beams. Unfortunately, the acquired signal was not sufficiently strong relative to the scattered background radiation from the prisms, beam splitters, and associated optics. Instead, the signal (which includes both the DFWM signal plus the linear background) was measured for different amounts of input power in each of the probe and pump beams. The presence of DFWM then appeared as a cubic departure from linearity when the two quantities, power out versus power in, were plotted against one another.

The results are shown in Fig. 2. The straight line drawn through the data represents the linear background component, and the curved line was arrived at by a least squares fit of the data to a third order polynomial, $Y = c_0 + c_1 X + c_3 X^3$. The background can be subtracted off from such data, and the remaining averaged DFWM signal-only portion of the data can be fit to a cubic-only dependence. This is shown in Fig. 3 for several experiments and the cubic dependence of the DFWM signal on incident power is clear. Each curve represents a different trial taken under different experimental conditions, i.e., different coupling efficiencies, detector gain, etc.

The magnitude of the conjugate signal can be calculated as outlined in Ref. 11. Specifically, it can be shown that P_4, the peak power of the reflected conjugate beam, is proportional to the product of the powers of the probe (P_3) and pump beams (P_1, P_2):

$$P_4 = D^{NL} \frac{L^2}{H^2} P_1 P_2 P_3.$$

The nonlinear coupling coefficient D^{NL} is proportional to the square of the appropriate nonlinear susceptibilities $\chi^{(3)}$, while H and L are the width and length of the interaction region which are determined by the beam widths and geometries. Estimating P_4 for our experimental conditions (detector sensitivity, coupling efficient, etc.), we obtained a value of 0.16μW (which corresponds to 0.01 pJ per pulse at 10 pulses per second), for a guided wave input in the nonlinear medium of 150 W (corresponding to 3 μJ per input beam). This is in reasonable agreement with the measured value of 0.1 μW based on the dominant reorientational contribution to the nonlinear susceptibility of CS$_2$. The thermal contribution to the nonlinearity, estimated from the CS$_2$ absorption coeffi-
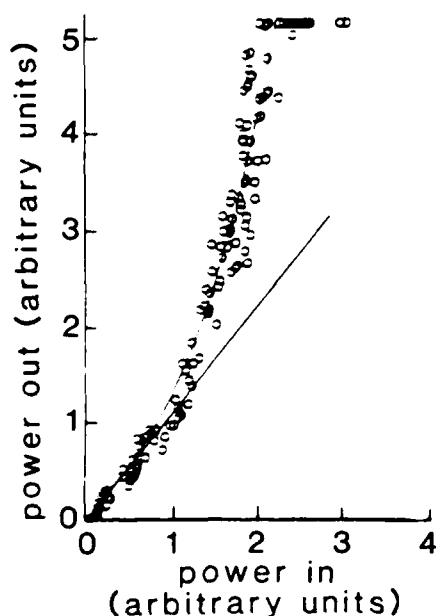

FIG 2 Signal power (P_4) vs incident power ($P_1 = P_2 = P_3$) with both the probe and pump beams present. The straight line represents the linear background and the curved line to the best least squares fit to the data.
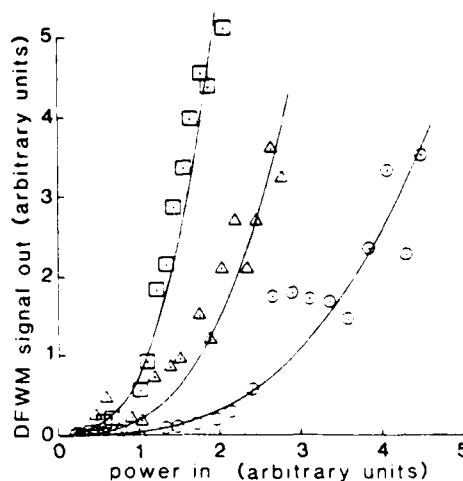

FIG. 3. DFWM data for three different experiments with the background component subtracted out. The curves drawn represent the best cubic fit.

cient, its thermal capacity, temperature dependence of refractive index, etc., was found to be negligible.

No measurable signal was obtained in the absence of the CS_2. This confirmed that the nonlinear mixing occurred in the nonlinear cladding medium, in agreement with calculations which included contributions from the glass film as well as the CS_2.

The major difficulties which we encountered were related to the relatively low DFWM cross section obtained with CS_2 used in a cladding geometry, of the order of 10^{-9}. For the available laser powers in the waveguide this resulted in stray light signal levels comparable to the DFWM signal, especially emanating from the probe-signal prism coupling region. We expect that the use of more highly nonlinear materials such as PTS[12] (bis-[P-toluene sulphonate] of 2,4-hexa-diyne-1,6-diol) for the film itself will lead to useful DFWM signals for all-optical signal processing.

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Ultrafast carrier and grating lifetimes in semiconductor-doped glasses

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The time dependence of both the photoluminescence due to carrier recombination and the gratings created by degenerate four-wave mixing were measured in semiconductor-doped color-filter glasses. Values ranging from 80 to less than 16 ps (laser pulse width limited) are measured in several different samples at various excitation levels. A slower mechanism, believed to be thermal in nature, is also observed with a lifetime in excess of 9 ns.

Materials with large and fast third order nonlinearities are of considerable interest for many applications in all-optical signal processing. Color-filter glasses consisting of a host glass doped with small semiconductor ($CdS_x Se_{1-x}$) crystallites have been reported by Jain and Lind[1] to have large third order susceptibilities ($\chi^{(3)} \simeq 10^{-8} - 10^{-9}$ esu). They performed degenerate four-wave mixing (DFWM) experiments with ~10-ns duration Q-switched laser pulses near the absorption edge of the color-filter glasses and noted a pulse width limited response time of their DFWM signal. They also measured the photoluminescence and found an upper limit of 1 ns for the carrier recombination time, limited by their detector response. In this letter we report both photoluminescence decay and degenerate four-wave mixing experiments on semiconductor-doped filter glasses with picosecond duration laser pulses. Our preliminary results indicate free-carrier (and grating) lifetimes ranging from 80 to less than 16 ps (laser pulse width limited).

The photoluminescence measurements were made using a mode-locked train of pulses of 30-ps duration and 50-μJ energy from a frequency-doubled Quantel neodymium: yttrium aluminum garnet (Nd:YAG) operating at 10 Hz. The time dependence of the photoluminescence from a range of color-filter glasses was measured with a Hamamatsu streak camera with 10-ps resolution. cw measurements indicated the photoluminescence spectrum to peak at wavelengths longer than the excitation wavelength so filters were used to block the 532-nm radiation from the detector.

A typical photoluminescence decay is shown in Fig. 1. For smooth, symmetric input laser pulses, the leading edge of the signal is identical to that of the input laser pulse, implying photoluminescence rise times limited by the 16-ps laser pulse rise time. The decay time of the photoluminescence is a measure of the excited carrier lifetime and Table I lists measurements of this decay time for several color filters. All of the glasses listed in Table I are strongly absorbing at the 532-nm excitation wavelength. The range of measured decay times for a specific glass results from measurements being made at different pump intensities. For example, a Corning 3-67 filter has decay times of 80 and 27 ps at pump intensity levels of 5 and 40 MW/cm³, respectively. This intensity-dependent decay would be expected from a carrier-dependent recombination mechanism such as Auger,[2] or from photoinduced surface recombination due to the small size of the crystallites (100–1000 Å) in which the free-carrier plasma is generated by interband absorption. We did not find any systematic change in the relaxation time with color-filter

band gap over the range of filters investigated. Since the relaxation time will depend strongly on the crystallite size, then the range of values measured is probably accountable for by variations in the fabrication process from filter to filter. The relaxation times may actually be reduced by controlling the crystallite size. An extensive qualitative study would be required to determine the functional form of this intensity-dependent lifetime and will be carried out in the future.

The degenerate four-wave mixing experiments were performed with the same frequency-doubled mode-locked Nd:YAG laser with variable delay lines inserted into the two pump beam paths to measure the effective lifetime of the photoinduced gratings. The probe beam was incident at an angle of a few degrees to the front pump beam. A DFWM signal was clearly observed in a Corning 3-68 filter (the same as used in Ref. 1). With all three input pulses coincident on the sample, a reflection efficiency of 1% at $\simeq 1$ MW/cm² pump power was observed. This signal level is slightly lower than that previously reported in Ref. 1, probably due to the poorer spatial quality of the unfiltered beams used in our experiments.

The DFWM signal obtained as a function of the delay of the backward pump pulse which is deflected by the grating formed by the simultaneous arrival of the front pump and probe pulses, is shown in Fig. 2. There are clearly two contributions to the observed DFWM signal. The regions some 30 ps before and 60 ps after the peak of the DFWM
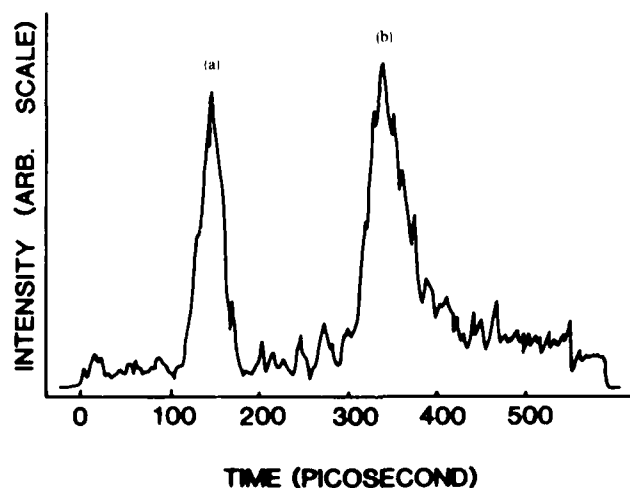

FIG. 1. (a) Incident laser pulse profile and (b) the photoluminescence decay for a Corning CS 3-68 color filter obtained with excitation at 532 nm.

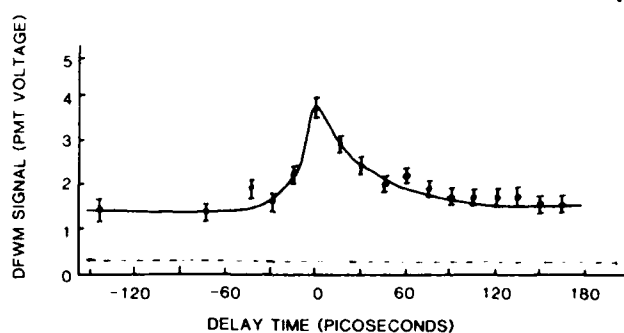| Glass		Decay time (ps)	
Corning	CS 3-68	16	~55
	CS 3-67	~27	~80
	CS 3-66	16	~32
	CS 2-63	16	~33
	CS 2-61	16 -	~27
	CS 2-59	16	~22
	CS 2-58	16	~33
Schott	OG 550	16	~33
	OG 570	16	
	OG 590	16 -	~22
	RG 645	16	~27

FIG. 2. Degenerate four-wave mixing signal as a function of delay time for a Corning CS 3-68 color glass filter. The dashed line corresponds to the background level when one of the pump beams is blocked.

signal are related to the generation and relaxation respectively, of the signal due to the induced free-carrier plasma. The short rise time of this signal indicates a turn-on time for this process limited by the laser pulse rise time of 16 ps, in agreement with the photoluminescence result. The grating decay time of ~30 ps (after correction for pulse width convolution broadening) is a measurement of the photoinduced free-carrier lifetime and is also in agreement with the photoluminescence measurements.

Underlying this fast process, a background grating contribution was found which persisted between the mode-locked pulses which were ≃9 ns apart. These long-lived gratings are probably thermal in nature and appear to contrast with Jain and Lind who observed a decay of their DFWM signal limited by their 8-ns laser pulse width. However, with their temporal resolution it would be difficult for them to differentiate between two relaxation rates, one faster than and one comparable to their laser pulse duration. We therefore conclude that the observed DFWM signals arise from contributions due to both a short-lived grating produced by interband absorption in the crystallites plus a background grating, probably thermal in nature due to bulk heating of the material. It may be possible to remove the effect of this thermal grating by choosing a host glass with better heat dissipation. Alternatively, one could utilize the fact that the "diffusion-free" electronic nature of these glasses allows the use of the small period "fast" grating created by the probe and back pump beam to generate the DFWM signal by judicious choice of polarizations of the input beams. The thermal contribution from such a small period grating would probably be significantly reduced. It may perhaps also be possible to produce the free-carrier excitation by two-photon absorption thus eliminating the background absorption. All of these alternatives are currently under investigation.

In summary, we have measured the lifetimes of photo-generated carriers for semiconductor-doped color-filter glasses. We measured, both by photoluminescence and DFWM, relaxation times of the nonlinearity to vary from 80 ps to less than 16 ps depending on excitation conditions. We also noted that there is a relatively long-lived background contribution, which it should be possible to considerably reduce to make these glasses excellent candidates as highly nonlinear materials for optical singal processing requiring fast excitation and recovery.

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Waveguide Fabrication in Nonlinear Semiconductor-doped Glasses

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Abstract

We have fabricated low-loss planar and channel waveguides in semiconductor-doped glasses which exhibit large third-order susceptibilities ($n_2 \sim 10^{-13}$ -10^{-14} m^2/W) with picosecond response times over a wide range of visible wavelengths.

Summary

Color-glass filters doped with small crystallites of CdS_xSe_{1-x} have been shown[1,2] to have a large third-order susceptibility corresponding to an intensity dependent index of refraction coefficient $n_2 = 10^{-13}$ -10^{-14} m^2/W. Thus these materials are potentially very useful for all-optical signal processing applications such as optical bistability, degenerate four-wave mixing (DFWM), optical logic, etc. This large nonlinearity has also been shown[2] to have characteristic response times for both excitation and recovery (which is not the case with most bulk semiconductor materials) of a few tens of picoseconds or less.

These glasses are commercially available as long wavelength pass optical filters ranging from yellow (for $x > 0.8$) to deep red ($x < 0.2$) corresponding to a gradual decrease in the bandgap with increasing selenium concentration.

DFWM experiments were performed with a frequency doubled Nd:YAG laser[1,2] to establish the magnitude of the nonlinearity which has been attributed to the generation of a short-lived electron-hole plasma in the individual crystallites. The lifetime of the photoexcited gratings was measured by introducing a delay arm into one of the pump beams of the DFWM set up and was composed of two components.[2] One component had a fast response of a few picoseconds attributed to the lifetime of the electron-hole plasma and the second, slower component, had a characteristic decay time of tens of nanoseconds which is attributed to a thermal mechanism. The response time was also measured by photoluminescence measurements[2] in a large number of color-filter glasses with 30 psec duration pulses and exhibited a range of response times from ~ 80 psec down to < 16 psec (instrument limited). Since the response times are expected to depend on the composition and size of the semiconductor crystallites, and on the density of excited carriers, such a range of results is only to be expected.

Glassses are an ideal candidate for passive integrated optical components for reasons such as low-loss, immunity to optical damage, compatability with fiber interfacing, etc. Integrated optical glass waveguides can be fabricated by deposition

or ion-exchange, with ion-exchange being the more flexible technique for making single-mode and multimode planar and two-dimensional channel devices. Sodium ions present in common glasses such as borosilicates and soda-limes have a high mobility and exchange well with monovalent alkali ions such as Tl^+, Cs^+ Ag^+ Rb^+ or K^+ all of which yield an increase in the refractive index of glass when exchanged with Na^+[3].

Detailed research into Ag^+/Na^+ ion-exchange fabrication of planar and channel waveguides and devices such as coherent couplers, interferometers and ring resonators has yielded successful results.[4,5,6] An attempt was therefore made to produce Ag^+/Na^+ ion-exchange waveguides in commercially available color filter glasses from a variety of manufacturers.

The initial attempt to obtain waveguides were unsuccessful but subsequent investigation into the composition of these commercially available borosilicate glasses indicated that there was less than 0.5% (by weight) of sodium present in these glasses. Subsequently we were able to obtain modified versions of these color-filter glasses in a soda-lime glass host with a sodium content of ~ 9.5% (standard soda-lime glasses used for ion-exchange have a sodium content of ~ 14%).

Using the higher sodium content glasses we again attempted to produce planar Ag^+/Na^+ ion-exchange waveguides. Samples of color-filter glass were cleaned and immersed in a $AgNo_3$ solution at temperatures ranging from 215-250°C and times from 15→ 60 minutes. The waveguide characteristics were measured by prism coupling of a HeNe laser which indicated considerable in-plane scattering and high loss. We attribute this scattering loss to aggregation and nucleation of silver atoms into collodial crystals. This effect has been observed wherever minor constituents such as sulphide, arsenic, antimony and ferrous ions are present in the glass.[3] Post production annealing of the Ag^+/Na^+ exchanged guides appeared to improve this scattering loss but not significantly enough to produce usefully low-loss guides.

Potassium and sodium (K^+/Na^+) ion-exchange has been shown[3,7] to be a very successful process for fabrication of waveguides with lower loss (< 0.2 dB/cm) than silver exchange and a lower index increase (Δn~ 10^{-3} as opposed to 10^{-2} for silver), because of the slower rate of diffusion of K^+ ions. Thus we attempted K^+/Na^+ ion-exchange in the high sodium content color-filter glasses by immersing them into molten KNO_3 at ~ 375°C for various times. By again prism coupling a HeNe laser, the guides produced were observed to be of good optical quality with very low scattering loss. The guided beams could be easily out coupled using a second prism and a strong centralised spot centered on a very weak scattered m-line was observed.

A series of channel structures were produced with lateral dimensions ranging from 100 μm down to < 3μm by standard photolithographic techniques and subsequently ion-exchanged in KNO_3 to produce single depth mode guides which were also single lateral mode in the 3 μm wide channels. Fig. 1 shows a series of 3 μm wide channel guides and Fig. 2 shows the guided streak of a He-Ne laser end-fire coupled into one of these guides. We have also used a cw mode-locked and Q-switched, mode-locked frequency doubled Nd:YAG laser operating at 532 m to end-fire couple into single mode channel waveguides and directional coupler structures in the color-filter glasses.

It should be noted that we were able to damage the 3 μm wide channel waveguides with a power density of ~ 1GW/cm^2 (Fig. 3). This power level is, however, significantly above the 1MW/cm^2 level where the large nonlinear effects were measured.[1] The trade off between absorption and nonlinearity as a function of laser frequency is obviously an important issue in the use of these materials for all-optical nonlinear integrated optics and is under study.

Wor' is currently in progress to produce optimised nonlinear optical channel wavegu : devices which will utilise the large and fast nonlinearity measured in these glasses in structures such as nonlinear directional couplers[8,9] Mach-Zender interferometers[10] and nonlinear ring resonators[11].

In conclusion we have successfully fabricated low-loss channel waveguides in semiconductor-doped glasses which, because of their fast response and large nonlinearity may be ideal materials for realisation of many proposed all-optical nonlinear integrated optical devices.
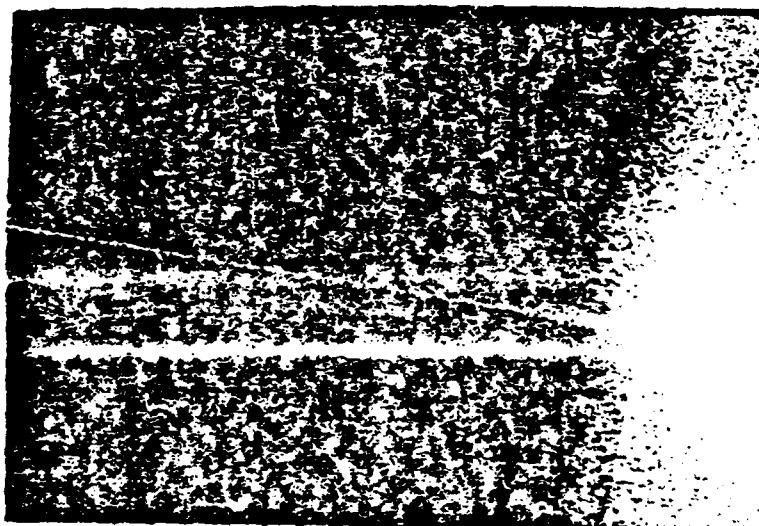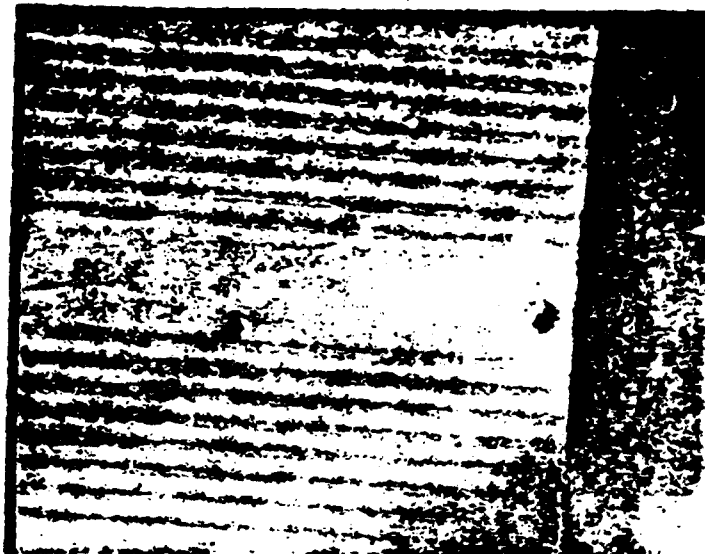
Figure Captions

Fig. 1. Contrast microscope photograph of 3 μm wide K^+/Na^+ ion-exchanged channel waveguides in semiconductor-doped color filter glass.

Fig. 2. Photograph of HeNe laser guided light in a 3 μm wide channel waveguide in semiconductor-doped glass.

Fig. 3. Phase contrast microscope photograph of optical damage created by Q-switched, mode-locked pulses from a frequency doubled Nd:YAG laser in a 3 μm wide channel arm of a directional coupler in semiconductor doped glass (power density ~ IGW/cm^2)

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